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Impact of air-borne or canopy-derived dissolved organic carbon (DOC) on forest soil solution DOC in Flanders, Belgium

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HIGHLIGHTS

- Soil solution DOC concentrations in Flanders forests increased (2002–2012).
- Soil solution DOC fluxes showed less systematic trends.
- Deposition could not explain the increase of soil solution DOC concentrations.
- Stand deposition had a rather limited impact on soil solution DOC.

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ABSTRACT

Dissolved organic carbon (DOC) in the soil solution of forests originates from a number of biologically and/or biochemically mediated processes, including litter decomposition and leaching, soil organic matter mineralization, root exudation, mucilage and microbial activity. A variable amount of DOC reaches the forest floor through deposition, but limited information is available about its impact on soil solution DOC. In this study, trends and patterns of soil solution DOC were evaluated in relation to deposition of DOC over an 11-year period (2002–2012) at five ICP Forests intensive monitoring plots in Flanders, northern Belgium. Trend analysis over this period showed an increase of soil solution DOC concentrations for all observed depth intervals. Fluxes of DOC increased in the organic layer, but were nearly stable in the mineral soil. Annual leaching losses of DOC were higher in coniferous (55–61 kg C ha^{-1}) compared to deciduous plots (19–30 kg C ha^{-1}) but embody less than 0.05% of total 1-m soil organic C stocks. Temporal deposition patterns could not explain the increasing trends of soil solution DOC concentrations. Deposition fluxes of DOC were strongly correlated with soil solution fluxes of DOC, but their seasonal peaks were not simultaneous, which confirmed that air-borne or canopy-derived DOC has a limited impact on soil solution DOC.

1. Introduction

Dissolved organic carbon (DOC) plays an important role in the C cycle in forest ecosystems and contributes to the sequestration of C in mineral soils (e.g. Neff and Asner, 2001; Buurman and Jongmans, 2005). It is a highly dynamic soil organic C pool and facilitates the transport and/or bioavailability of nutrients and pollutants, such as nitrogen (N), phosphorus, sulphur and trace metals (Qualls et al., 1991; Kalbitz et al., 2000). It also forms a major pathway for C transfer from terrestrial to aquatic ecosystems, and provides a significant indirect source of CO_2 emission to the atmosphere (Freeman et al., 2001, 2004; Cole et al., 2007).

During the past decennia DOC concentrations and/or fluxes increased in many forests, peatlands, streams and lakes of northern and central Europe and eastern North America, while stable or decreasing DOC trends in these areas were less frequently observed (Freeman et al., 2001, 2004; Monteith et al., 2007; Lindroos et al., 2008; Oulehle and Hruška, 2009; Wu et al., 2010; Löfgren and Zetterberg, 2011; Couture et al., 2012; Akselsson et al., 2013). Besides to climate change, rising DOC levels have been attributed to changes in soil solution chemistry induced by the overall decline of sulphur depositions, suggesting that ecosystems are recovering towards their high-DOC, preindustrial state (de Wit et al., 2007; Oulehle and Hruška, 2009; Borken et al., 2011; Evans et al., 2012). A decline of acidifying depositions repeatedly leads to a decrease in soil solution ionic strength and pH increase, which both enhance

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DOC solubility in forest soils (Kalbitz et al., 2000; Vanguelova et al., 2010; Graf Pannatier et al., 2011; Evans et al., 2012; Kerr and Eimers, 2012; Verstraeten et al., 2012). The main source of DOC in the soil solution of forests are a number of biologically and/or biochemically mediated processes, including plant and root litter decomposition and leaching, soil organic matter mineralization, root exudation, mucilage and microbial activity (Kalbitz et al., 2000; Yano et al., 2000; Hansson et al., 2010). The main factors explaining soil solution DOC levels are tree species and soil type. Generally higher DOC levels are recorded in coniferous compared to deciduous forests, primarily due to more intensive leaching of DOC from an evergreen canopy, the higher C:N ratio of needles compared to leaves and the higher mass of the forest floor in coniferous forests (Currie and Aber, 1997; Borken et al., 2011; Arisci et al., 2012). Highly podzolized soils tend to have a lower DOC retention capacity than less acidified soils and may exhibit unusually high DOC outputs (Guggenberger and Zech, 1993).

Soil solution DOC levels display a characteristic seasonal pattern and a depth pattern in the soil profile. Peak concentrations are observed near the second half of the growing season in response to higher soil temperatures, while the highest C losses coincide with elevated water fluxes during the winter period (Buckingham et al., 2008; Sleutel et al., 2009; Wu et al., 2010; Futter et al., 2011; Gielen et al., 2011). Concentrations and fluxes of DOC generally decrease with depth, due to translocation from the organic layer towards the mineral B-horizon, where C is removed from soil solution mainly by abiotic processes, like precipitation as organo-metal complexes and/or by adsorption to solid Fe- and Al-phases (Guggenberger and Zech, 1993; Michalzik et al., 2001; Jansen et al., 2005; Fuss et al., 2011).

A considerable amount of DOC reaches the forest floor through atmospheric deposition or canopy leaching, but limited information is available about its role in the forest C cycle and its impact on soil solution DOC (Yano et al., 2000; Michalzik et al., 2001; Sleutel et al., 2009). In this study, we examined the contribution of air-borne or canopy derived DOC to concentrations and fluxes of soil solution DOC and their seasonal patterns using long-term monitoring data.

Long-term monitoring of deposition and soil solution concentrations provides valuable data to evaluate changes in soil solution chemistry. Flanders is participating in the ICP Forests programme (www.icp-forests.org), launched in 1985 under the Convention on Long-Range Transboundary Air Pollution (CLRTAP) of the United Nations Economic Commission for Europe (UNECE, www.unece.org). The objectives of this study are: 1) to evaluate trends and patterns of soil solution DOC concentrations and fluxes and 2) to assess the impact of air-borne or canopy-derived DOC on fluxes, concentrations and seasonal patterns of soil solution DOC.

2. Materials and methods

2.1. Study area

Five plots of the ICP Forests intensive monitoring network (Level II) in Flanders, northern Belgium, were included in this study (Fig. 1, Table 1). Belgium has a moderate Atlantic climate with an annual precipitation of 852 mm and a temperature of 10.5 °C (long-term averages for 1981–2010, www.meteo.be). Plots were circular with an area of 0.25 ha. Two plots are located in coniferous forest: *Pinus sylvestris* L. in Brasschaat (BRA) and *Pinus nigra subsp. laricio* (Poiret) Maire in Ravels (RAV). Both stands are situated in the northern Campine ecoregion on a sandy soil (Arenosol) with a C:N ratio of 30–35 in the organic layer (mor humus). The soil profile in RAV is well-drained, while in BRA the infiltration of water is locally slowed down by clay lenses at 50–125 cm depth. Three other plots are installed in deciduous forest: *Fagus sylvatica* L. in Wijnendale (WIJ) and Hoeilaart (HOE), a mixture of *Quercus robur* L. and *F. sylvatica* L. in Gontrode (GON). WIJ is located in the western part of Sandy Flanders, on a sandy loam soil (Umbrisol). HOE is located in the Loess belt of Flanders, on a silt to silt loam soil (Alisol) with deep groundwater table (>30 m). GON is located on the slopes of a brooklet valley in the Dender-Klein Brabant ecoregion, on a silt loam to loam soil (Stagnosol). The deciduous stands have an organic layer C:N ratio of 22–25 (moder humus). A more detailed description of stand and soil characteristics, land-use history and differences in pollution climate of these plots was given in Verstraeten et al. (2012).

2.2. Sample collection

Samples of soil solution and deposition were collected fortnightly over the period studied (2002–2012), according to the guidelines of the ICP Forests manual (Hansen et al., 2013; Nieminen et al., 2013). Soil solution from the organic layer was sampled with four (RAV, BRA, GON, HOE) or six (WIJ) randomly located zero tension lysimeters per plot. They consisted of a five cm high stainless steel box covered with a nylon mesh, installed just below the forest floor. Soil solution from the mineral soil was sampled with ceramic cup suction lysimeters (Eijkelkamp) at three random locations per plot. Each location was equipped with two to four lysimeters at each of three depths in the capillary zone (topsoil, subsoil and deeper mineral soil). The lysimeters were pressurized to 0.6 bar using a vacuum pump two days before sampling. Samples

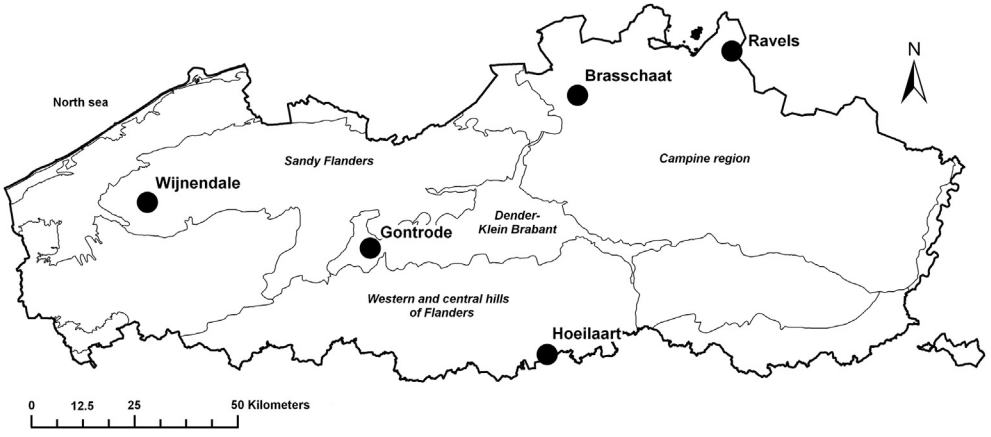

Fig. 1. Location of the five intensive monitoring plots in relation to ecoregions in Flanders (after Sevenant et al., 2002).

Table 1
Characteristics of five intensive monitoring plots in Flanders. Mean annual temperature (MAT) and precipitation (MAP) are long-term averages (1981–2010, Royal Meteorological Institute of Belgium, www.meteo.be). Soil and humus types were defined in 2007 (IUSS Working Group WRB, 2006, 2007; Zanella et al., 2011). Total organic carbon stock is given for the forest floor (FFC) and the upper 1 m of the mineral soil (SOC) (De Vos, 2009).

Plot	Coordinates		Elevation	MAT	MAP	Tree species	Planting	Soil type	Humus	FFC	SOC	Rooting	Groundwater
	N	E	m	°C	mm		Year		Type	ton ha^{-1}	ton ha^{-1}	Depth (cm)	Range (m)
Coniferous forests													
RAV	51°24′07″	05°03′15″	35	10.4	887	*Pinus nigra subsp. laricio* (Poiret) Maire	1930	Endogleyic Hyperfolic Hypobrunic Albic Arenosol	Mor	52.2	149	185	1.5–2.5
BRA	51°18′28″	04°31′11″	14	10.8	882	*Pinus sylvestris* L.	1929	Endogleyic Hypobrunic Albic Hypoluvic Arenosol	Mor	38.7	82.5	160	1.2–2.3
Deciduous forests													
WIJ	51°04′11″	03°02′14″	31	11.0	867	*Fagus sylvatica* L.	1935	Endogleyic Hyperfolic Umbrisol	Mor	64.3	200	170	0.9–2.3
GON	50°58′31″	03°48′15″	26	10.6	786	*Quercus robur* L., *Fagus sylvatica* L.	1918	Luvic Folic Stagnosol	Moder	40.3	132	180	1.5–1.8
HOE	50°44′45″	04°24′47″	129	10.7	854	*Fagus sylvatica* L.	1909	Glossalbic Hypocutanic Alisol	Moder	20.4	93.7	195	>30

were bulked to one composite sample per depth per plot at every sampling event, using the entire collected volume.

Bulk precipitation was sampled with four bulk collectors, located in the open field at less than 500 m distance from each plot. Bulk collectors consisted of a polyethylene funnel (14 cm Ø) placed at 1 m height, and connected to a subterranean 2 L polyethylene bottle. A nylon mesh (1 mm^2) was placed in the funnel to avoid contamination by large particles. Throughfall was sampled in each plot with ten systematically distributed bulk collectors of the same type as open field collectors. At every sampling event, the volume collected by each bulk collector was determined and samples were bulked together to one sample for precipitation and throughfall per plot. Stemflow sampling was conducted only for *Fagus*, since stemflow for *Pinus* and *Quercus* was shown to be negligible during a preceding testing period. At each deciduous stand five trees of different size (mean diameter, $\pm 1 \times$ and $\pm 2 \times$ standard deviation of the initial tree population in 1994) were selected. Stemflow collectors consisted of flexible polyvinylchloride collars/gutters (7 cm Ø) attached horizontally to the stem at 1 m height, draining to a series of 200 L polyethylene storage containers mounted in a cascade system. The volume collected by each individual tree was determined at every sampling event. Subsamples were taken from all full containers, with subsample volumes weighted to tree diameters, and bulked together. Stemflow volumes, obtained from individual sample trees, were upscaled on area basis using information of basal area.

Table 2
Seasonal Mann-Kendal trends for water fluxes (mm), DOC concentrations (mg l^{-1}) and DOC fluxes (kg ha^{-1} y^{-1}) for bulk and stand precipitation, organic layer and mineral soil layers (2002–2012), with mean annual values, letters (a–e) indicating groups of plots with comparable mean, Sen's slope (annual change) and significance (*: $p < 0.05$, **: $p < 0.01$, ***: $p < 0.001$).

Plot	Depth (cm)	Water fluxes		DOC concentrations		DOC fluxes	
		Mean	Slope	Mean	Slope	Mean	Slope
Bulk precipitation							
RAV		923a		2.2a	+0.1***	20.2a	
BRA		960a		2.4a	+0.1*	19.0a	
WIJ		929a		2.2a	+0.1**	19.0a	+0.5*
GON		838a		2.0a	+0.1***	15.0a	
HOE		971a		1.9a	+0.1***	17.0a	+0.8**
Stand precipitation							
RAV		649ab		17.5a		106.2a	
BRA		770a		14.9a		102.8a	
WIJ		718ab		7.9c		49.0b	
GON		621b		11.2b		60.0b	
HOE		766a		7.8c		49.6b	
Organic layer							
RAV		513ab		41.9a	+0.9*	211.5a	+7.0*
BRA		590a		35.4b	+2.0***	201.3a	+5.6*
WIJ		644a		27.0c	+1.9***	185.1a	+10.7***
GON		431b		35.5b	+1.0**	160.2a	
HOE		578a	+17.5*	33.5b		189.6a	
Topsoil							
RAV	10–25	395a		46.4a	+0.9**	185.4a	+7.0**
BRA	15–25	348a		38.1b	+1.3***	131.7b	
WIJ	10–20	355a	−13.5*	35.0c	+1.1***	118.7bc	
GON	10–20	328a		25.8d	+0.4**	84.5d	
HOE	10–15	398a	−10.2*	22.6e	+0.6***	88.0cd	
Subsoil							
RAV	30–45	295ab	+13.1**	42.6a	+0.6*	121.5a	+6.0***
BRA	3–55	264b		31.0b		76.5b	
WIJ	45–70	253b		19.1c	+0.6***	48.9cd	
GON	25–40	314ab	−10.6*	19.7c	+0.5***	57.5bc	
HOE	20–30	372a		8.0d	+0.5***	28.7d	+1.0*
Deeper mineral soil							
RAV	70–95	297ab	+11.9**	19.6b	+0.5***	61.3a	+3.9***
BRA	70–90	268b	−15.5***	21.2a	+0.8***	54.5a	
WIJ	75–110	189c	−6.5**	10.4d	+0.4***	19.3b	
GON	45–55	208c		13.2c	+0.5***	27.8b	
HOE	35–55	367a		8.1e	+0.6***	29.7b	+1.4***

2.3. Chemical analysis

Samples were treated and analysed as prescribed by the ICP Forests manual (Hansen et al., 2013; Nieminen et al., 2013). Quality control included analysis of control samples (blanks, reference

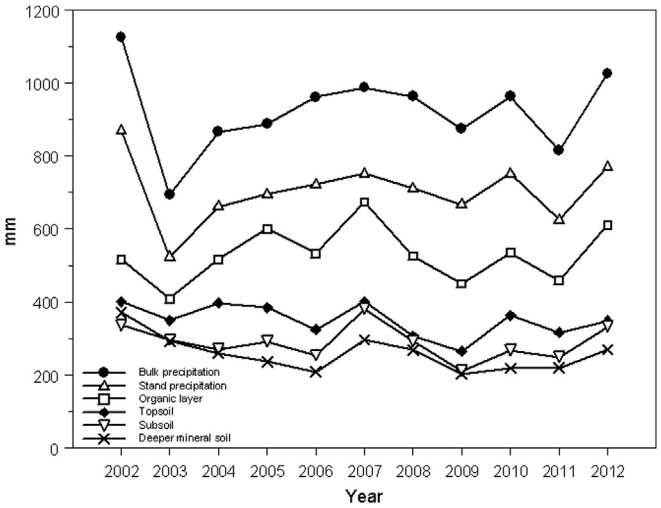

Fig. 2. Mean annual bulk precipitation, stand precipitation (throughfall + stemflow) and soil water flux per depth for the five plots (mm).

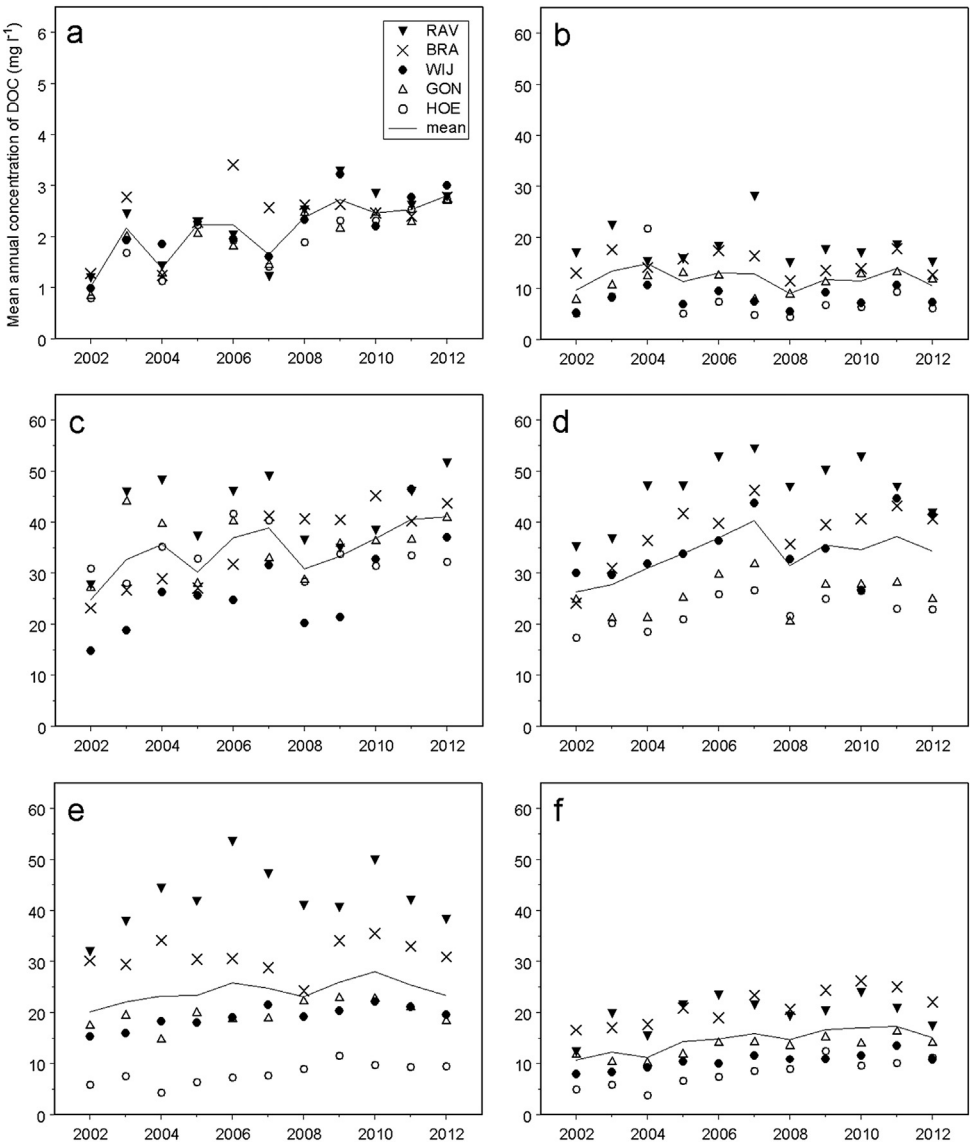

Fig. 3. Mean annual DOC concentration (mg l^{-1}) in bulk precipitation (a), stand precipitation (b) organic layer (c), topsoil (d), subsoil (e) and deeper mineral soil (f).

material, replicates) and participation in the yearly ICP Forests water ring tests. Water samples (500 ml subsamples of each collected fraction) were kept cool during transport, filtered (0.45 μm), stored in darkness at 4 °C and analysed within 48 h after sampling. Concentrations of DOC (mg l^{-1}) were determined using a TOC-analyser (Shimadzu TOC 5050A, limit of quantification 0.1 mg l^{-1}). Concentrations of Na^+ (mg l^{-1}) were determined using ion chromatography (IC, limit of quantification 0.1 mg l^{-1}).

2.4. Data handling

All field data (e.g. sample mass) and analytical results were subjected to a detailed quality check and validation procedure, according to the guidelines of the ICP Forests Working Group on Quality Assurance/Quality Control (König et al., 2013). Deposition of DOC (kg ha^{-1}) was calculated as the product of DOC concentrations (mg l^{-1}) and the collected volume (l m^{-2}) of bulk precipitation, throughfall or stemflow. Stand deposition was calculated as the sum of throughfall and stemflow deposition.

Because water balance models have not yet been calibrated for the plots, vertical water fluxes were estimated for each depth using the sodium (Na^+) mass balance approach, assuming conservative behaviour of Na^+ in the ecosystem and negligible weathering of Na^+ from the mineral soil (Bailey et al., 2003). Soil solution DOC fluxes for the fortnightly sampling periods were calculated as the product of water flux and DOC concentration. Monthly soil solution DOC fluxes were calculated as the sum of the two fortnightly fluxes; annual fluxes as the sum of the twelve monthly fluxes. Because a number of data were missing for several evident reasons (e.g. no sampling performed due to snow, or not enough sample volume available for analysis) this would result in an underestimation of DOC fluxes. In order to obtain more realistic DOC fluxes, missing DOC concentrations were interpolated using a logarithmic curve for all measured DOC concentrations during the same year in function of sampling period number. If not enough data were available to obtain a reliable curve ($R^2 < 0.2$), DOC concentrations were interpolated as the average of the previous and the following measured DOC concentration. Missing Na^+ concentrations were interpolated as the average of the previous and the following measured Na^+ concentration. For soil solution DOC concentrations measured in a period without rainfall, no water flux could be calculated. In that case the DOC flux was

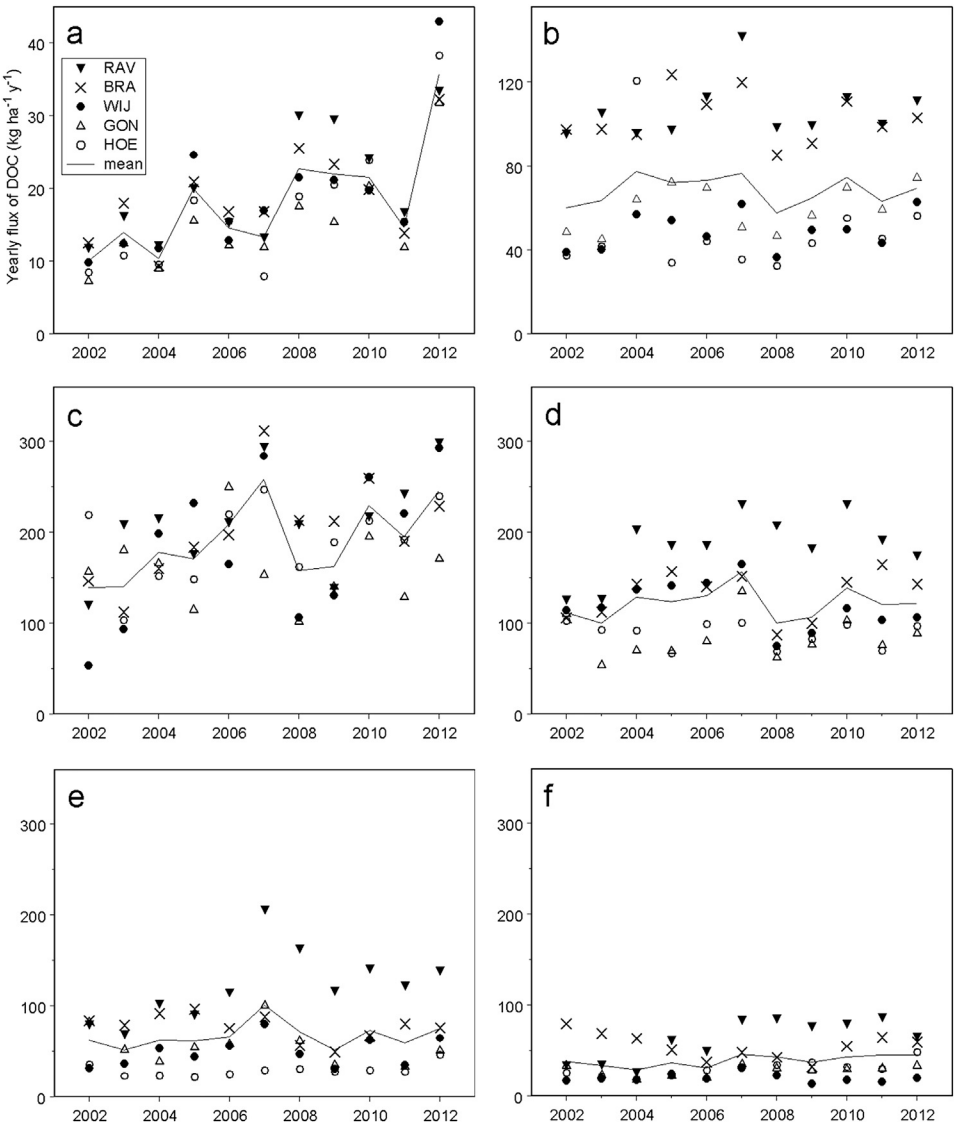

Fig. 4. Annual flux of DOC (kg ha^{-1} y^{-1}) for bulk precipitation (a), stand precipitation (b) organic layer (c), topsoil (d), subsoil (e) and deeper mineral soil (f).

interpolated as the average of the previous and the following DOC flux obtained from measured DOC concentrations, if possible weighted by DOC concentration and sample volume. We assumed that DOC fluxes through the deeper mineral soil represented C losses below the rooting zone.

2.5. Statistical analysis

The nonparametric Seasonal Mann Kendall Test (Hirsch and Slack, 1984) was applied to detect monotonic trends in monthly aggregated data for water fluxes, DOC concentrations and DOC fluxes. Trends were evaluated by the significance of Sen's slope (Sen, 1968). The change (increase or decrease) of DOC concentrations and fluxes was expressed as a percentage in function of the mean value for the 11-year observation period and Sen's slope:

$$\text{change}(\%) = \frac{11 \times \text{slope}}{\text{mean} - \frac{1}{2}(11 \times \text{slope})} \times 100$$

Cross-site statistics were performed by means of ANOVA/Tukey's range test. The impact of airborne or canopy-derived DOC on soil solution DOC was evaluated by comparing single DOC fluxes in stand precipitation with fluxes in soil solution, using Spearman's rank correlation ('cor.test' function in *R*) and simple linear regression ('lm' function in *R*), the latter after natural log transformation of DOC fluxes to obtain normally distributed residuals (R Core Team, 2012). Seasonal variation within the data was tested in R by introducing harmonic terms (sine and cosine) in a stepwise regression of the dependent variable *y* versus time (e.g. Thimonier et al., 2008), in the form:

$$y = c_1 + c_2\sin t + c_3\cos t + c_4\sin 2t + c_5\cos 2t + c_6\sin 3t + c_7\cos 3t + c_8\sin 4t + c_9\cos 4t + c_{10}\sin 5t + c_{11}\cos 5t$$

where $t = 2\pi/365 \times$ (day of the year) and $c_1 - c_{11}$ are regression coefficients. Sine and cosine terms up to 5 *t* were included, which allows the model to show processes which go up or down within a little more than one month. This makes the analysis sensitive enough to reveal the important features of the annual cycle, but not too sensitive to single irregularities.

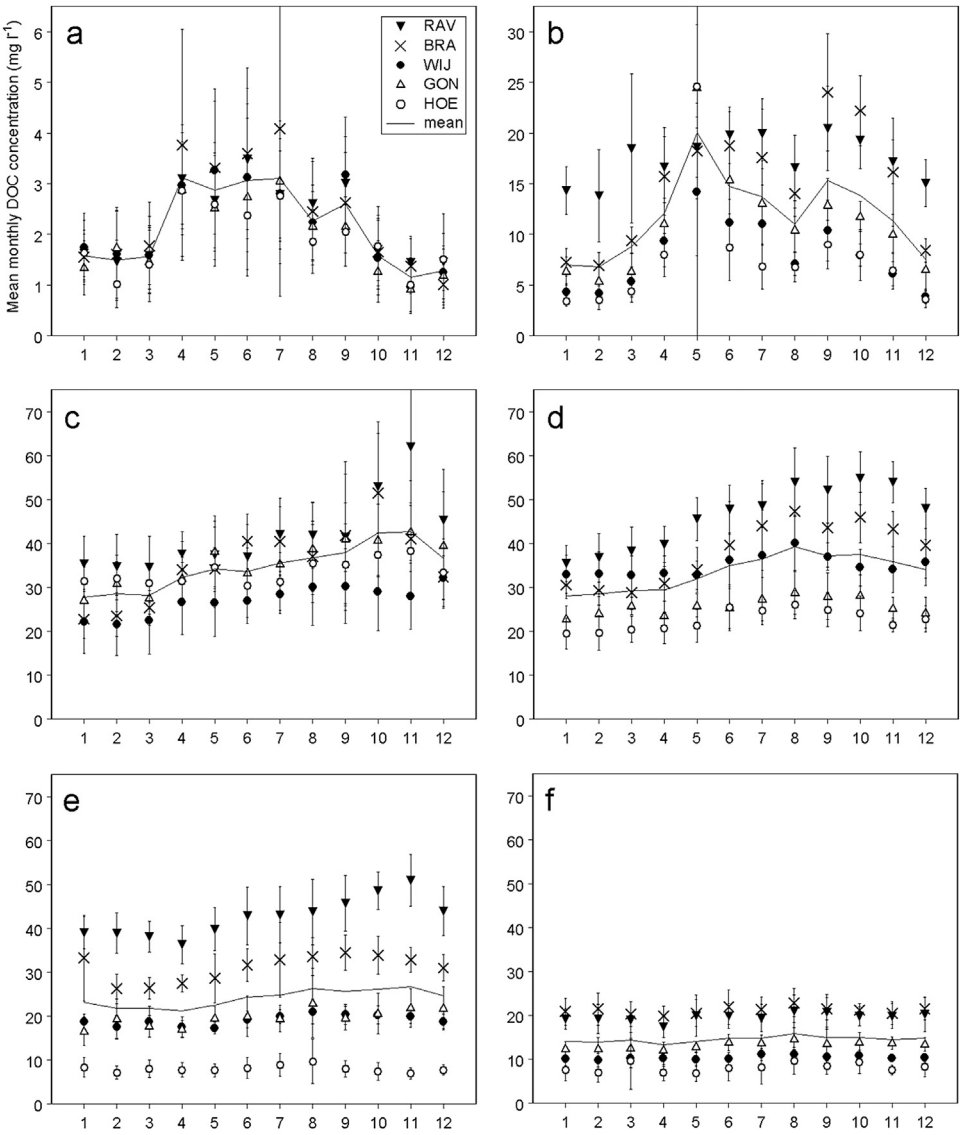

Fig. 5. Mean monthly DOC concentration (mg l^{-1}) in bulk precipitation (a), stand precipitation (b), and soil solution of the organic layer (c), topsoil (d), subsoil (e) and deeper mineral soil (f) (2002–2012). Error bars represent the 95% confidence interval for the original data (before interpolation).

3. Results

3.1. Long-term trends of DOC

Trends and significances for annual water fluxes, monthly DOC concentrations and DOC fluxes are presented in Table 2. The annual precipitation showed substantial year-to-year variation, with wet years in 2002 and 2012 and dry years in 2003 and 2011 (Fig. 2). In spite of this, annual bulk precipitation and stand precipitation showed no significant trend between 2002 and 2012. Soil water fluxes showed a significant trend at several individual soil layers, but there were no indications for a consequent increase or decline of the soil water flux through the soil profile at any of the plots.

Bulk precipitation DOC concentrations significantly increased at the five plots during the observation period, by 36–110% (Fig. 3). Bulk precipitation DOC fluxes significantly increased at WIJ and HOE by 31% and 67% respectively, but remained stable at the other plots (Fig. 4). Stand precipitation DOC concentrations and fluxes did not change significantly at any of the plots.

In the soil solution, DOC concentrations remained stable in the organic layer at HOE and the subsoil at BRA (Fig. 3). Elsewhere, DOC concentrations significantly increased by 26–130% in the organic layer, 17–44% in the topsoil, 16–103% in the subsoil and 32–146% in the deeper mineral soil. Soil solution DOC fluxes through the organic layer remained stable in GON and HOE, but significantly increased by 36–93% at the other three plots (Fig. 4). Soil solution DOC fluxes through the mineral soil remained stable at BRA, WIJ and GON, but increased significantly in RAV and HOE (except the topsoil at HOE).

3.2. Within and across-site variability of DOC

Soil solution DOC concentrations and fluxes generally decreased with depth, with an exception for the plots on sandy soils (RAV, BRA and WIJ), where mean annual DOC concentrations were higher in the mineral topsoil than in the organic layer (Table 2). Concentrations and fluxes of DOC for stand precipitation and soil solution were higher in coniferous than in deciduous plots, and this difference became increasingly important with increasing soil depth. Stand precipitation DOC fluxes were nearly twice as high in coniferous compared to deciduous plots. Accordingly, leaching losses of DOC below the rooting zone were

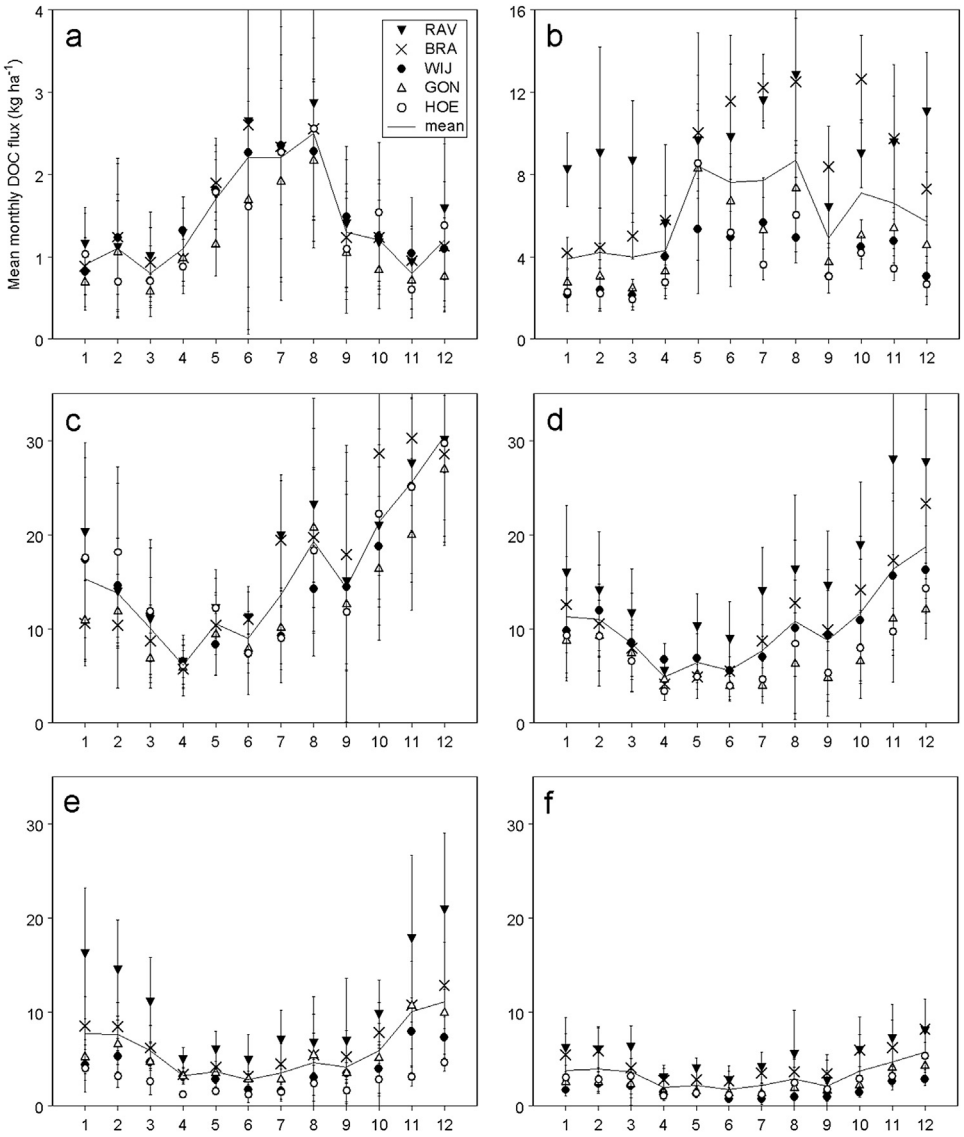

Fig. 6. Mean monthly DOC flux (kg ha^{-1}) for bulk precipitation (a), stand precipitation (b), and through the organic layer (c), topsoil (d), subsoil (e) and deeper mineral soil (f) (2002–2012). Error bars represent the 95% confidence interval for the original data (before interpolation).

significantly higher in coniferous (55–61 kg C ha^{-1} y^{-1}) compared to deciduous plots (19–30 kg C ha^{-1} y^{-1}). These C losses embody 0.12–0.14% and 0.03–0.15% of the C stock in the forest floor and 0.04–0.07% and 0.01–0.03% of the organic C stock in the upper 1 m of the mineral soil in deciduous and coniferous plots respectively (Table 1). We found no correlation between C losses and soil organic C stocks.

3.3. Impact of air-borne or canopy-derived DOC

Bulk precipitation DOC fluxes amounted to 9–10% of the estimated annual DOC fluxes through the organic layer, while stand precipitation DOC fluxes amounted to 50–51% and 26–37% of the estimated annual DOC fluxes through the organic layer of coniferous and deciduous plots respectively.

Stand precipitation DOC fluxes were strongly ($p < 0.001$) correlated with soil solution DOC fluxes at all depths in each plot. The linear models showed that 23–24% of the variance in soil solution DOC fluxes could be explained by stand deposition (Fig. 7). The slope of regression lines showed little difference between the soil depths (0.50–0.58), while the intercept declined with depth from 1.32 to −0.36, reflecting the removal of DOC by mineralization and sorption towards the deeper mineral soil.

Soil solution DOC concentrations and fluxes showed a clear seasonal pattern at all locations, with the highest mean monthly values observed near the end of the growing season, particularly from August till December (Figs. 5, 6 and 8). The seasonal variation of DOC concentrations gradually declined with depth, to become no longer significant in the deeper mineral soil, while water fluxes and DOC fluxes showed seasonality at all depths (Table 3). Bulk and stand precipitation DOC fluxes were highest during late spring and summer (May–August). The seasonal pattern of stand precipitation DOC fluxes was most eminent in the deciduous plots, where DOC concentrations showed two peaks: a first one in May around the moment that buds break and leaves are unfolded, and a second one in September just before the start of litterfall.

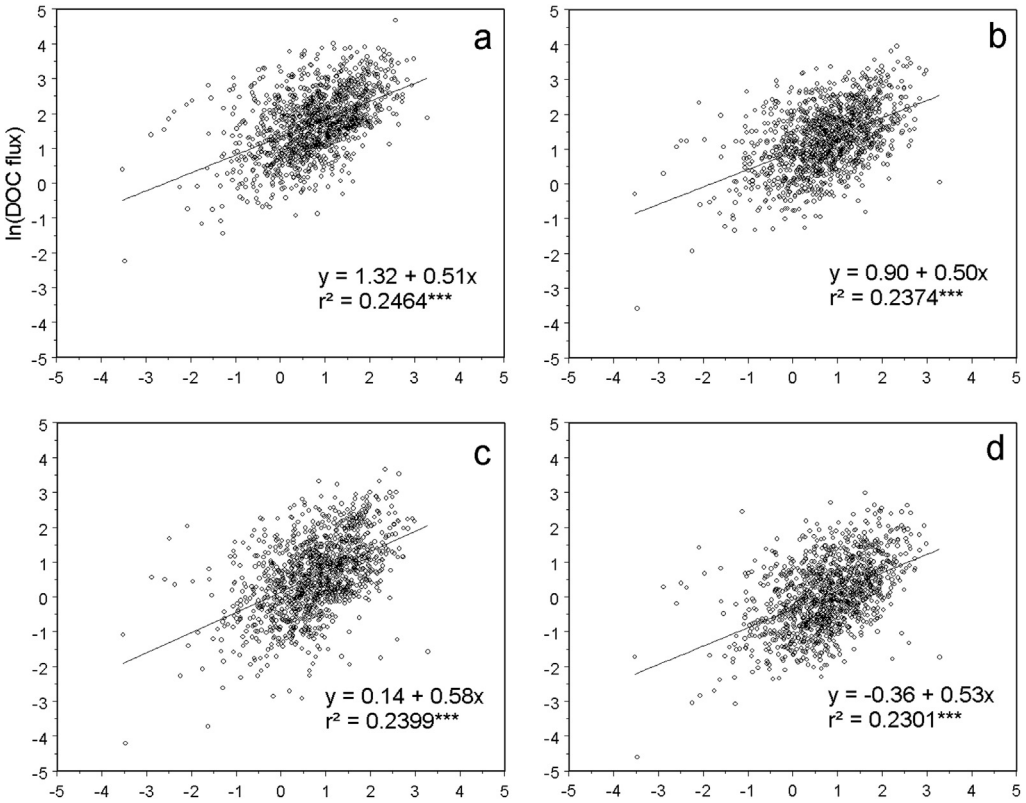

Fig. 7. Linear models for log transformed DOC fluxes (kg ha^{-1}) in soil solution of the organic layer (a), topsoil (b), subsoil (c) and deeper mineral soil (d) in function of stand precipitation.

4. Discussion

4.1. Uncertainties in the calculation of water and DOC fluxes

The sodium mass balance approach could result in an underestimation of soil water fluxes if there is a considerable Na^+ weathering flux. For the plots studied, we may assume that Na^+ weathering is negligible, regarding the minor amounts of Na-Feldspar and mica in the finer soil textures and the absence of that mineral in the sandy soils (Van Ranst et al., 2002). However, soil solution DOC fluxes could still be slightly underestimated, because ceramic lysimeter cups tend to adsorb DOC (Menéndez et al., 2003).

4.2. Long-term trends of DOC

The observed increase of soil solution DOC concentrations over the last ten years is in agreement with the recent predominance of increasing trends in soil solution DOC observed in Europe and North America (Freeman et al., 2001, 2004; Monteith et al., 2007), although some studies reported stable or negative trends, particularly at high northern latitudes (e.g. Wu et al., 2010; Akselsson et al., 2013). The increase of DOC can be basically explained by recovery processes in the soil and soil solution initiated by the overall decline of sulphur depositions (de Wit et al., 2007; Borken et al., 2011; Evans et al., 2012). Past acid conditions have reduced litter decomposition rates, allowing a pool of relatively labile organic matter to accumulate, from which DOC may be generated when acidity decreases (Oulehle and Hruška, 2009). Because atmospheric sulphur depositions and ion concentrations in the soil solution showed a strong and parallel decline at the five plots between 1994 and 2010, the increase of soil solution DOC concentrations can at least partly be explained by recovery from acidification (Verstraeten et al., 2012). The increase of soil solution DOC at the five plots appears to be unrelated to air-borne or canopy-derived DOC, since stand precipitation DOC concentrations and fluxes remained stable during the observation period.

In the soil solution, DOC fluxes showed less systematic trends than concentrations, which may be explained by the strong dependence of DOC fluxes on water fluxes and the relatively high year-to-year variation in precipitation and soil water fluxes (Neff and Asner, 2001; Buckingham et al., 2008; Gielen et al., 2011). This could imply that longer time series (>11 years) are recommended to study trends in soil solution DOC fluxes.

The concentrations and fluxes of DOC observed at the five plots are in the order of magnitude reported by other studies in temperate forests in Europe (Buckingham et al., 2008; Sleutel et al., 2009; Wu et al., 2010; Borken et al., 2011; Löfgren and Zetterberg, 2011; Arisci et al., 2012). Carbon leaching losses of up to 61.3 kg ha^{-1} y^{-1} were recorded, but embody less than 0.05% of measured total 1-m soil organic C stocks (De Vos, 2009).

4.3. Within and across-site variability of DOC

The observation that soil solution DOC fluxes, and consequently C losses below the rooting zone, were higher in coniferous compared to deciduous plots, accords with the results of similar studies and could be explained by the interaction between tree species, soil type and pollution climate (Currie and Aber, 1997; Borken et al., 2011; Arisci et al., 2012). Regarding the tree species, DOC levels are generally higher in coniferous forests, primarily due to more intensive leaching of DOC from an evergreen canopy, the higher C:N ratio of needles compared to leaves and the higher mass of the forest floor (Arisci et al., 2012; van den Berg et al., 2012). Regarding the pollution climate, the higher DOC levels in coniferous plots could be related to the higher N deposition in these plots (Guggenberger and Zech, 1993; Sleutel et al., 2009;

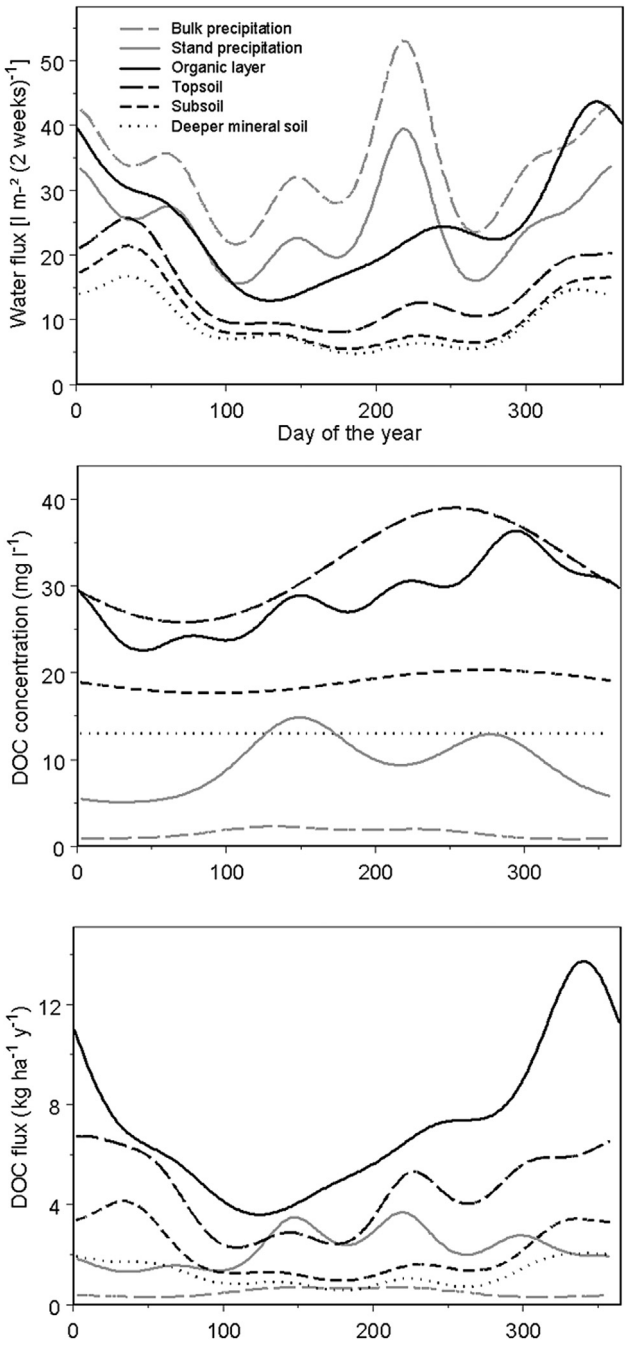

Fig. 8. Modelled seasonal variation of water fluxes, DOC concentrations and DOC fluxes for bulk precipitation, stand precipitation, organic layer, topsoil, subsoil and deeper mineral soil.

Verstraeten et al., 2012). Regarding the soil type, soils in the deciduous plots have a higher DOC retention capacity, due to their higher clay content, cation exchange capacity (CEC) and base saturation, providing more sorption sites and higher potential for cation bridging of organic compounds (Guggenberger and Zech, 1993; Kerr and Eimers, 2012).

4.4. Impact of air-borne or canopy-derived DOC

The strong correlation that we found between stand precipitation DOC fluxes and soil solution DOC fluxes could be explained by their strong dependence on the amount of precipitation, as observed earlier in temperate forest ecosystems in North America and Europe (Michalzik et al., 2001; Neff and Asner, 2001; Buckingham et al., 2008). On the other hand we found that seasonal peaks of DOC in stand precipitation and soil solution did not occur simultaneous, which was also observed in Norwegian forests (Wu et al., 2010). This confirms that the impact of deposition is limited, and that soil solution DOC originates mainly from biologically and/or biochemically mediated processes in the soil compartment, including the activity of soil biota, soil respiration, root development and root exudation (Yano et al., 2000; Neff and Asner, 2001; Carrara et al., 2004; Buckingham et al., 2008; Futter et al., 2011).

However, several studies point out that the impact of air-borne or canopy-derived DOC could increase in the future (Vitasse et al., 2009; Pitman et al., 2010; Clark et al., 2009; Reyer et al., 2013). In the canopy, the activity of phytophagous insects – especially aphids generating honeydew – can increase DOC levels during summer (Michalzik and Stadler, 2005; Pitman et al., 2010). With the progress of climate change, the intensity of insect attacks is expected to increase, with a possibly more severe impact on the forest C cycle (Clark et al., 2009; Pitman et al., 2010). Climate change also extends the length of the growing season in temperate regions, and according to climate change scenarios may increase the net primary production (CO_2 effect) of forests in Northern Europe (Vitasse et al., 2009; Reyer et al., 2013).

We suggest that the impact of DOC deposition on soil solution DOC concentrations and fluxes will be further examined in the future, preferentially by means of long data series from a larger number of sites within a broader geographical extent, e.g. at the European level. Experimental studies, for example with addition of DOC containing labelled carbon (C^{14}), may provide new insights into this matter.

Table 3
Regression models for the seasonal variation of water fluxes, DOC concentrations and DOC fluxes for bulk and stand precipitation, organic layer and mineral soil layers, with the number of observations (n), R^2 and significance (*: $p < 0.05$, **: $p < 0.01$, ***: $p < 0.001$).

	Water fluxes			DOC concentrations			DOC fluxes		
	n	R^2	Sig.	n	R^2	Sig.	n	R^2	Sig.
Bulk precipitation	1206	0.08	***	1206	0.12	***	1206	0.09	***
Stand precipitation	1206	0.08	***	1206	0.20	***	1206	0.11	***
Organic layer	1024	0.20	***	1026	0.07	***	1024	0.18	***
Topsoil	777	0.20	***	782	0.14	***	777	0.17	***
Subsoil	854	0.23	***	871	0.01	*	854	0.19	***
Deeper mineral soil	843	0.20	***	861	0.004		843	0.16	***

5. Conclusions

Concentrations of DOC in the soil solution of forests in Flanders increased between 2002 and 2012 at all plots, while DOC fluxes showed less systematic trends. These trends could not be explained by temporal deposition patterns. Concentrations of DOC and annual C leaching losses below the rooting zone were higher in coniferous compared to deciduous plots but embody less than 0.05% of total 1-m soil organic C stocks. Our results confirmed that air-borne or canopy-derived DOC has a limited impact on soil solution DOC, and that soil solution DOC originates mainly from biologically and/or biochemically mediated processes in the soil compartment.

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